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ABSTRACT

Currently, most of our thinking about the defects responsible for initiating laser damage considers them as featureless absorbers. However, an increasing body of evidence, particularly involving multi-wavelength irradiation, suggests electronic structure of damage initiators is important in determining both initiation and conditioning behaviors in KDP. The effective absorption coefficient of energy under multi-wavelength irradiation cannot be accounted for by a structureless absorber, but is consistent with an initiator with a multi-level structure. We outline the evidence and assess the ability of such a simple multi-level model to explain these and other experimentally observed behaviors

Keywords: KDP/DKDP, laser damage, damage initiation

1. INTRODUCTION

The importance of KDP/DKDP frequency conversion crystals has stimulated significant investigation into the initiation of bulk damage and the improvement in damage susceptibility by pre-exposure to laser light below damage causing levels (conditioning). Considerable progress has been made in improving the damage susceptibility of such material. Thus far, it has not been possible to unambiguously identify the initially absorbing precursors that lead to damage in KDP. Such identification is desirable both from a scientific viewpoint and with a practical view of optimizing means of reducing damage incidence.

A significant database of experimental damage behaviors in KDP/DKDP is being accumulated, which will help, constrain the possible candidate initiators. The purpose of the present paper is to consider a detailed absorber model that is capable of accounting for at least some of these behaviors. Among these interesting behaviors, the following may be noted.

The threshold laser energy fluence has been noted to scale as a power of pulselength near ½ for many materials. In KDP, the bulk damage threshold has been found[1-3] to scale with pulselength τ roughly as $\tau^{0.24\text{-}0.35}$ for wavelengths of 355 and 532 nm. Pulselength scaling for conditioned material[4] is similar to that for unconditioned material. If the damage process were controlled by only the magnitude of the electric field, one would expect the damage threshold fluence to scale linearly with the pulselength. On the other hand, if only sufficient energy were required, one would expect the threshold fluence to be independent of pulselength, at least over a significant range of pulselength. The actual observed pulselength scaling has generally been taken to indicate a dissipative process, i.e. energy being absorbed by the damage precursor is offset against energy being lost by, e.g., heat conduction. A credible model of damage initiation has to account for the observed pulselength dependence.

Not only does the threshold fluence vary with pulselength, but so does the resultant damage site size[1, 5] with the size being roughly proportional to pulselength, at least for pulselengths of 1 ns and larger. Experiments[6] also indicate a damage size that varies proportional to the time delay between pump and probe pulses. These results suggest a pressure pulse whose duration is proportional to that of the laser pulse determines the size scale. These very interesting results will not be used in describing damage initiation.

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The wavelength dependence[7] of the damage initiation fluence shows a general increase with wavelength over the range of 300-550 nm with the very interesting addition of two "steps" in threshold fluence at apparent sub-multiples of the bandgap energy. This result suggests a connection between the electronic structure of the damage initiating precursor and that of the surrounding host crystal. This connection is the subject of the model given below.

When the crystal is subject to laser radiation at both 2ω and 3ω (2nd and 3rd harmonic of 1.06 μm light), the resultant pinpoint damage density is found[8] to depend on a linear combination of the fluences at the two wavelengths. This behavior is taken to indicate that it is the total absorbed energy that matters and that the two wavelengths have slightly different absorptivities. The 2ω fluence is equivalent to a certain amount of 3ω fluence and the two-color situation is described by an effective 3ω fluence corresponding to the total absorbed energy. The two color damage density results from ref. [8] are shown in Fig. 1. The individual curves may be collapsed onto one curve by shifting each to the right an amount dependent on the 2ω fluence and plotted in terms of the effective 3ω fluence. This is shown in Fig. 2.

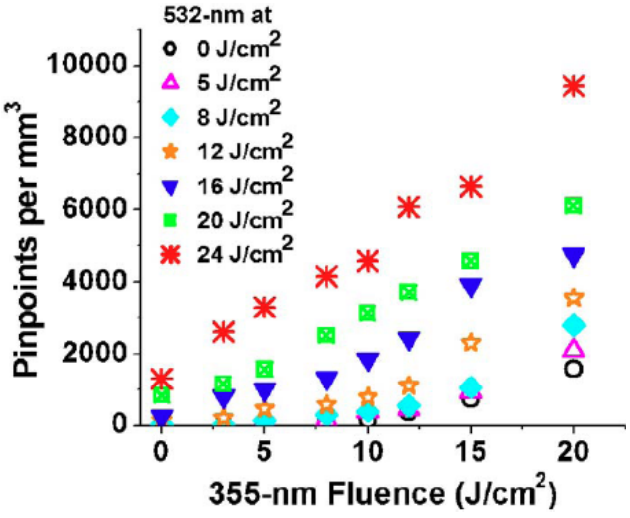

Fig. 1. Measured pinpoint density in DKDP as a function of 3ω fluence for various values of 2ω fluence. From. Ref. [8].

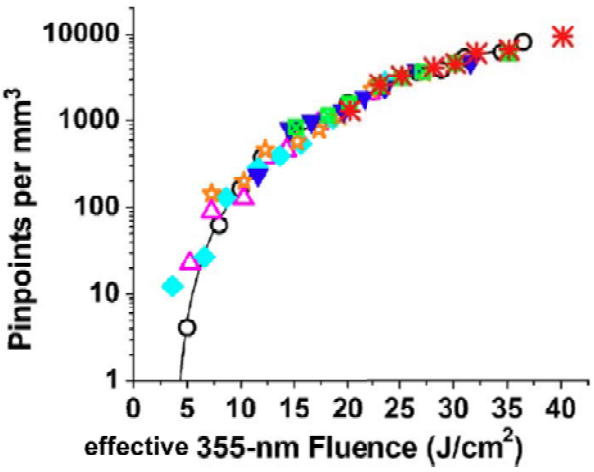

Fig. 2. Measured pinpoint density in DKDP as a function of effective 3ω fluence for various values of 2ω fluence from Fig. 1. From. Ref. [8].

At present, there is no one physically reasonable model that gives a good account of all the above behaviors. We have previously developed[9] a model of damage precursors as small featureless absorbers distributed in size. Such a model leads to a natural explanation of pulse-length dependence of the threshold fluence with an exponent near 0.5. A promising extension[10] of such an approach to include distributed absorptive is presently underway.

Direct application of the small absorber model predicts that the relative effectiveness of 2ω to 3ω light should be independent of laser fluence. Experiment[8] indicates some intensity dependence, which suggests the internal structure of the absorber, is important. Below, we present a simple model of a possible level structure of a KDP damage initiator and explore how such a model can account for the multi-color damage initiation and pulse length dependence behavior of KDP/DKDP.

2. MULTI-LEVEL MODEL.

The wavelength dependent threshold results suggest a level structure as shown schematically in Fig. 3 in which states 1 and 2 in the bandgap assist promoting electrons from the ground state to the conduction band Here the τ_i are relaxation times, the σ_{ij} are cross sections, the E_i are energy levels and I is the laser intensity. Although we do not know the

parameters directly, there are some hints. For example, elegant experiments[11] have shown the lifetime of conduction band electrons in fused silica to be about 1 ps and we assume that value here. The pump-probe experiments suggest a lifetime τ_1 on the order of 1 ns and that is also assumed. The experiments suggest a 2ω bottleneck in transfer from level 1 to level 2 and we assume cross sections to reflect this. The system is described by a simple set of rate equations.

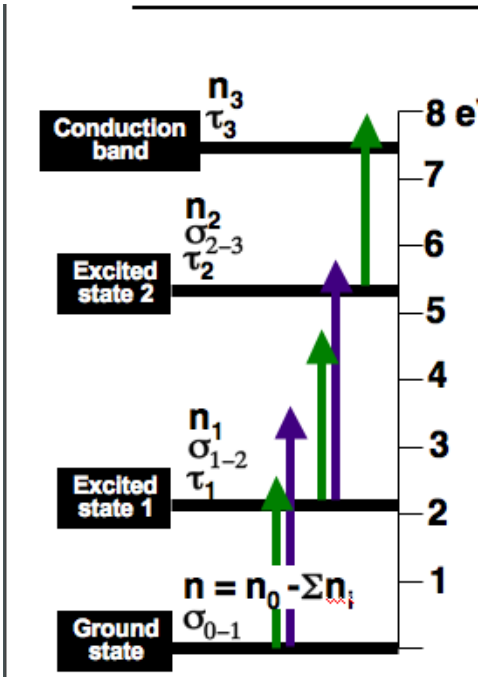

Fig. 3: Proposed energy level scheme for KDP bulk damage precursors. Green arrows refer to 2ω photons, purple arrows to 3ω photons.

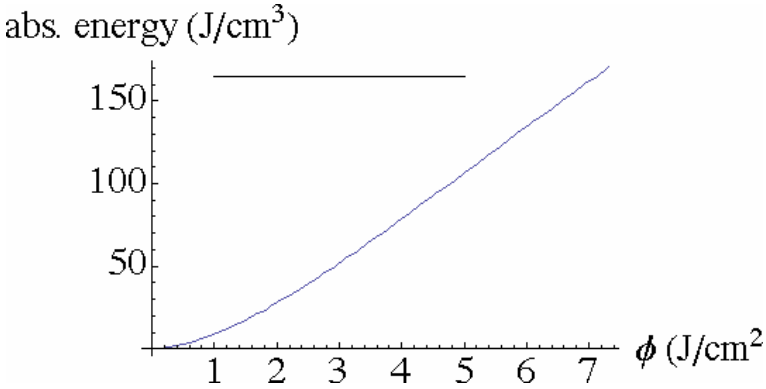

Fig. 4: Calculated absorbed energy as a function of incident fluence assuming crystal has to be heated to ~1000K to initiate damage. The line represents the damage threshold absorbed fluence. In this case, the threshold is about 7 J/cm^2, which is the proper order of magnitude.

$$\frac{dn}{dt} = \frac{n_1}{\tau_1} - \frac{\sigma_{01}}{E_1 - E_0} I \; n$$

$$\frac{dn_1}{dt} = -\frac{n_1}{\tau_1} + \frac{n_2}{\tau_2} + \frac{\sigma_{01}}{E_1 - E_0} I \; n - \frac{\sigma_{12}}{E_2 - E_1} I \; n_1$$

$$\frac{dn_2}{dt} = -\frac{n_2}{\tau_2} + \frac{n_3}{\tau_3} + \frac{\sigma_{12}}{E_2 - E_1} I \; n_1 - \frac{\sigma_{23}}{E_3 - E_2} I \; n_2$$

$$\frac{dn_3}{dt} = -\frac{n_3}{\tau_3} + \frac{\sigma_{23}}{E_3 - E_2} I \; n_2$$

The results shown below have assumed the following parameter values:

$\tau_1=10\tau_2=1000\tau_3 = 1$ns,

$$\sigma_{01}^{3\omega} = 2.3,\ \sigma_{12}^{3\omega} = 1.6,\ \sigma_{23}^{3\omega} = 22,\ \sigma_{01}^{2\omega} = 2.3,\ \sigma_{12}^{2\omega} = 0.32,\ \sigma_{23}^{2\omega} = 22$$
$$E_1 - E_0 = 2.3,\ E_2 - E_1 = 3.2,\ E_3 - E_2 = 2.2$$

Here the times are in ns, the energies in eV and the cross-sections are in arbitrary units.

We can test the order of magnitude of various quantities in the model for reasonableness. For example, we might suppose that the damage threshold corresponds to heating the material to some threshold temperature. We will take this to be 0.1 eV (about 1000K) for argument's sake. In this case, since one mole of KDP is 58.4 cm^3, this condition corresponds to an absorbed energy density of 164 J/cm^3. In our model, electrons dropping down from level to level can transfer energy to the lattice at a rate per volume of $\frac{n_1}{\tau_1}(E_1 - E_0) + \frac{n_2}{\tau_2}(E_2 - E_1) + \frac{n_3}{\tau_3}(E_3 - E_2)$. The necessary total precursor density, n_0, to reach the threshold absorbed energy in 2 ns is then found to be $n_0=1.5\text{x}10^{18}$ cm^{-3}. Assuming this value of n_0, we can also express the laser intensity and absorbed energy in physical units with the results shown in Fig. 4. The precise values in this figure are not important, but again the reasonable orders of magnitude argue for the reasonableness of the model.

3. RESULTS

Assuming flat in time pulses of 2ω and 3ω light, the rate equations above lead to steady state values of the level populations. The level populations determine the effective energy absorption rates for the two wavelengths. The relative effectiveness of 2ω compared to 3ω light, called γ in ref. [8], can then be calculated. In the experiments, a correlation was made between this effectiveness and the level of damage density ρ(ϕ) as shown in Fig. 5a. Our theoretical values can be compared by assuming a relation between fluence ϕ and density ρ. We chose a power law dependence of damage density on total absorbed energy fluence, $r\sim\phi^3$ and found the results shown in Fig. 5b. The two figures are qualitatively similar.

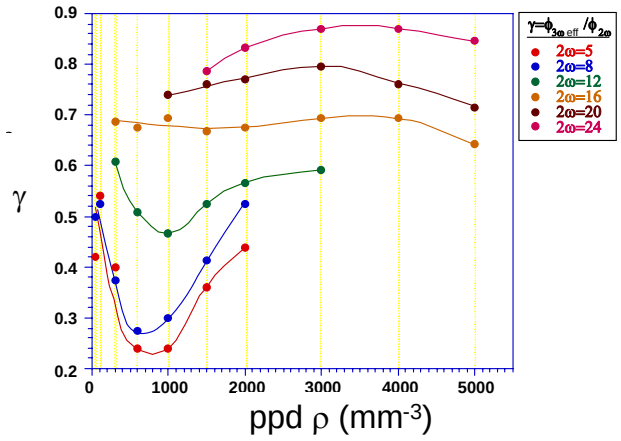

Fig. 5a:Experimental determination of relative effectiveness of 2ω and 3ω fluences as a function of pinpoint density at fixed values of 2ω fluence (ref [8])

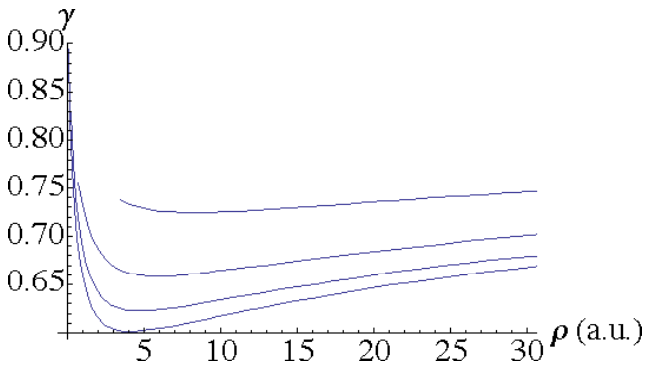

Fig. 5b: Theoretical determination of relative effectiveness of 2ω and 3ω fluences as a function of pinpoint density assuming power law dependence of damage density on total absorbed energy. Curves correspond to differing proportions of 2ω and 3ω fluences.

Let us turn now to the expected pulselength dependence of the damage threshold. If we assume that the threshold is determined by the absorption of a fixed energy density, the variation of the threshold incident fluence may be examined. As above, the effective absorption depends on the magnitude of the intensity. We have previously shown[9] how the absorbed energy is expected to change with size, absorptivity and laser fluence in a population of absorbers with differing sizes. The fluence ϕ needed to heat the absorber up to temperature T_x was found to be

$$\phi = \frac{4\kappa\tau T_x}{Qa}\left(1 - \exp\left[-\frac{4D\tau}{a^2}\right]\right)$$

where κ is the thermal conductivity, τ the pulselength, D the thermal diffusivity, a the absorber radius and Q the absorptivity. The threshold is set by the absorber that heats most efficiently, i.e. for the most efficient size which depends on pulselength. When the absorptivity Q itself depends on the laser intensity (or fluence), this equation becomes an implicit equation for the threshold fluence. Solving this equation as a function of laser pulselength reveals the pulselength dependence.

The present multi-level model does not lead to a general power law pulselength dependence of damage threshold over all pulselengths. However, the dependence may be well fit by a power law over some limited extent of pulselengths. Since this dependence has been established experimentally for KDP/DKDP only over a limited pulselength extent also, this is, perhaps, not a fatal flaw.

Carrying out this calculation leads to the result for absorbed energy shown in Fig. 6. The calculated absorbed energy rises slightly faster than linearly with incident intensity because of the intensity dependent effective absorption. This results in a predicted scaling of the damage threshold approximately as the 0.27 power of pulselength, which is comparable to the observed value.

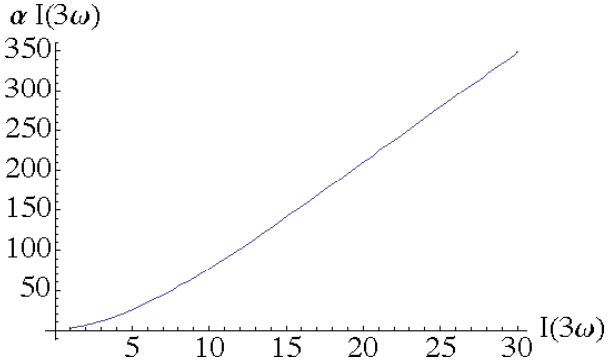

Fig. 6: Calculated absorbed energy rises slightly faster than linearly with incident intensity because of intensity dependent effective absorption.

4. SUMMARY

A simple multi-level model of the initiating precursors in KDP with level spacings, relaxation times and level to level transition rates selected to be consistent with known experimental behaviors gives a reasonable account of the fluence dependence observed for two color illumination. In this model, the total absorbed energy fluence is the important physical quantity. A pulselength dependence of the damage threshold similar to that reported experimentally can be obtained over a limited fluence range. This model deserves further development.

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